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While formally quantum computation has become a rather well defined theoretical science, physical demonstrations are few, limited to several qubits, and to several steps in processing. At present, there is not a recognized, scalable physical method of implementation that meets the promise of true computation. We have proposed nonlinear spectroscopic methods of coherence manipulation, and in particular, Time-Frequency Resolved Coherent Anti-Stokes Raman Scattering (TFRCARS) in the molecular re-vibronic Hilbert space as a novel scheme for implementing quantum logic. The method has advantages with its demonstrable massive parallelism, therefore well suited for information transfer, while its scalability remains unresolved, both theoretically and with regard to physical implementations. We are actively following the latter, with the concept of an inhomogeneously indexed array of single quantum centers as the physical realization.

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FINAL REPORT, 2004

QUANTUM COMPUTING & INFORMATION TRANSFER BY OPTICAL MANIPULATION OF MOLECULAR COHERENCES

AFOSR GRANT F49620-01-1-0449

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I. Accomplished Science

While formally quantum computation has become a rather well defined theoretical science, physical demonstrations are few, limited to several qubits, and to several steps in processing. At present, there is not a recognized, scalable physical method of implementation that meets the promise of true computation. We have proposed nonlinear spectroscopic methods of coherence manipulation, and in particular, Time-Frequency Resolved Coherent Anti-Stokes Raman Scattering (TFRCARS) in the molecular ro-vibronic Hilbert space as a novel scheme for implementing quantum logic. The method has advantages with its demonstrable massive parallelism, therefore well suited for information transfer, while its scalability remains unresolved, both theoretically and with regard to physical implementations. We are actively following the latter, with the concept of an inhomogeneously indexed array of single quantum centers as the physical realization.

In an extended paper,[1] we demonstrated experimentally that the elements sufficient for executing universal quantum logic, namely the set of one-qubit rotations and the two-qubit Controlled-Not (CNOT) gate, are naturally contained in TFRCARS. The experiments were carried out in room temperature molecular iodine, in which superpositions of 10^3 eigenstates are prepared, manipulated and read-out at the uncertainty limit, through the four-wave mixing process of TFRCARS. We demonstrated the universal gates by constructing a logical map between TFRCARS observables and input pulses in terms of manipulated multi-dimensional quantum coherences. The paper contains all of the important original concepts regarding nonlinear spectroscopy in Hilbert space as a viable approach to Q-computing. If not full fledged computing, then at least quantum communication and encryption can be expected as a near term practical application of our approach.

While defining how logic gates can be constructed in TFRCARS is the logical starting point, it is as useful to give concrete examples of how such gates can be assembled together to execute a well-defined algorithm. To this end, we illustrated how the Deutsch-Jozsa (DJ) algorithm,[2] which has become a benchmark for experimental demonstrations of quantum computing, could be implemented in TFRCARS.[3] The DJ algorithm demonstrates the advantage of the use of qubits in superposition, as opposed to classical definite bits, to gain exponential advantage in search algorithms. Our specific example showed how up to 8-qubit searches can be made with 99% distinguishability, with a single projective query. The latter

impressive limit is reached only with the use of shaped pulses, to counteract anharmonicity induced dispersion of the manipulated superposition both in the preparation and in the readout.

The design of the DJ algorithm brings out several important considerations, among which is the recognition that as the data base is increased the fidelity of the algorithm drops, unless optimally tailored laser pulses are used. Although this is not a surprise, it is useful to have quantitative estimates of fidelity and therefore success of pulse shaping. To this end, we have built a custom pulse shaper, using liquid crystal spatial light modulators (256 individually addressable liquid crystal channels). The pulse shaper was constructed, demonstrated, and operated with genetic algorithms to tailor the pulse to the desired shape. However, prior to the implementation of the shaper in the Q-search algorithm, the student responsible for the project was disabled (terminal illness). The project has been reassigned, the new student has redeveloped the system back to full operation, and we expect shaped-pulse demonstration of logical operations to be executed by the end of this summer, supported by a new AFOSR grant on the same subject.

The above demonstrations of proof of principle were carried out in the gas phase, where decoherence occurs on a time much longer than the manipulations. Practical implementations will undoubtedly occur in the solid state. As such, we have carried out the four-wave mixing measurements in the solid state, in rare gas matrices doped with molecular iodine. In the first ever of such measurements, we demonstrated that beside the elimination of rotations, electronic dephasing acts as a momentum filter to enable a much sharper definition of vibrational coherences.[4] A rigorous treatment of the anharmonicity effects,[5] and the potential contribution of inhomogeneity due to lattice imperfections,[6] were developed theoretically using self-consistent field treatment of vibrations and phonons. The latter analysis predicted the effects observed in the relatively soft solid of Ar, as verified through subsequent experiments.[7] Moreover, in the soft solid, the use of high peak power lasers to carry out the nonlinear manipulations was observed to lead to destruction of the cage and dissociation of the molecule. A complete analysis of cage breaking in Ar, through nonlinear optical excitation, was carried out in an experiment/theory paper.[8] These effects are completely negligible in the harder lattice of Kr, as the four-wave mixing measurements were transferred to Kr.[9] A rather complete map of vibrational dissipation and dephasing of $v = 1 - 19$ and at $T = 4K - 40$ K has been obtained in this system, as part of the Ph.D. thesis work of M. Karavitis,[10] and as training grounds for two postdoctoral fellows. A manuscript summarizing the complete work on this prototypical system is now in preparation.[11]

One possible scheme of scaling, is to go beyond four-wave mixing. We have recently demonstrated that in electronically resonant cases, coherent six-wave mixing can be executed and uniquely assigned.[12] This approach alone will be rather limited in scalability arguments. However, 6-wave mixing does provide for rather unique manipulations of coherences, as such can be relied upon for specific processing of information.

Decoherence is the process by which quantum systems decay into the classical world, and therefore provides the fundamental limitation to the scalability of Q-operations. In molecular systems, the mechanics of decoherence is not fully understood, and the subject of intense research. The six-wave mixing approach was specifically devised to prepare and interrogate vibrational coherences on the electronically excited state of the molecule. This enables a rather unique window in the observation of vibrational decoherence in the strong coupling limit, a limit where system-bath coherence can be expected to persist. We have given an explicit treatment of the observed vibrational decoherence, using semi-classical initial value

representation approach to explicitly simulate a lattice of 300 degrees of freedom. The results of the simulation are in excellent agreement with the experiments.[12] The published work suggests that rather generally, in the limit where the system communicates with the bath (transfers energy or phase, or both) on a time scale faster than the bath fluctuation periods, then quantum coherent evolution of system and bath must be expected. Indeed this could be demonstrated with rigor through 4-wave mixing measurements, by the preparation of two vibrational packets on the excited electronic state of a given molecule, and to observe sustained coherence in a system undergoing extensive dissipation.[13] It is possible to show that quantum coherent dissipation immediately implies a coherent bath, and to the extent that the activated bath consists of many nearly orthogonal degrees of freedom, quantum coherence in macroscopically distinguishable states (alternatively referred to as Schroedinger's cat) is achieved. We show that nearly 12 phonons of the bath are activated and remain simultaneously coherent for the duration that the system can be observed (~10 vibrational periods). In essence, a twelve qubit entanglement is demonstrated in the experiment. This is a rather impressive achievement, and points at the feasibility of useful 12 qubit manipulations in the solid state.

II. Publications

Beside the cited references, 10 manuscripts in print, one manuscript submitted (13), and one in preparation (11), material for several additional papers has been generated that we plan to publish in the near future.

III. Personnel

The two principal students that have carried out the referenced work, **Mike Karavitis** (experimental) and **Zsolt Bihary** (theoretical), have both completed their Ph.D. theses. Karavitis is presently employed in R&D at Intralase, while Bihary is carrying out a postdoctoral fellowship at Northwestern (with Prof. M. Ratner). The work on demonstrating Schroedinger's cat is carried out by **Darren Segale**, a Ph.D. student in his third year of studies. He is also the lead researcher on the pulse shaping optics. The shaper was originally assembled and operated with genetic algorithm control by **Mike Sgroi**, a Ph.D. student who had to drop out of the program due to illness.

Earlier contributions to the experiment and theory were spearheaded by Drs. **R. Zadoyan** and **Daniela Kohen**, respectively. We have also closely collaborated on theoretical concepts with **Prof. Dani Lidar** and his Ph.D. student **D. R. Glenn**, of the University of Toronto.

Noncolinear Optical Parametric Amplifiers (NOPAs) were constructed and extensively used in the accomplished work. These were built by students, with initial consultation with **Prof. N. Schwentner** of the Free university of Berlin who spent the summer of 2001 at UCI.

IV. Conference Presentation

The list of the presented invited lectures based on this work includes:

7/01 Gordon Conference on Coherent Control of atomic and molecular dynamics (Mt. Holyoke, Mass.)
Preparation, Manipulation, and Interrogation of molecular coherences, or is it reset, process and readout of the quantum register?

8/01 American Physical Society Meeting, (College Park, MD)
Time and Frequency Resolved Four-Wave Mixing Spectroscopy: From Quantum Control to Quantum Computing

10/01 Yerevan State University (Yerevan, Armenia)
Time and Frequency Resolved Four-Wave Mixing Spectroscopy: From Quantum Control to Quantum Computing

3/02 APS Conference (Indianapolis, IN)
Time-frequency Resolved Coherent Anti-Stokes Raman Scattering: Quantum Control to Quantum Computing

4/02 University of Michigan, Ann Arbor (Ann Arbor, MI)
Connecting Spectroscopic Observables to Underlying Molecular Dynamics

8/02 4th International Conference on Low Temperature Chemistry (Keuruu, Finland)
Time Resolved Spectroscopies in Matrices

10/02 Cryo-crystals (Munich)
Coherence manipulations through multilinear spectroscopy in condensed phase

1/03 Physics of Quantum Electronics (Utah)
Quantum Information transfer and control through multilinear spectroscopy

1/03 U. Wisconsin
The manipulation of molecular Coherences / Decoherence: new perspectives and prospects

3/03 U. of Oregon
The manipulation of molecular Coherences / Decoherence: new perspectives and prospects

5/03 Northwestern (*Colloquium*)
The manipulation of molecular Coherences / Decoherence: new perspectives and prospects

7/03 GRC, Electronic Spectroscopy (Bates College, ME)
Semiclassical treatment of nonlinear spectroscopy in condensed media - (given by M. Ovchinnikov)

10/03 UCI
Quantum coherences, decoherence, and the emergence of classicality

2/04 Third International Meeting on Photodynamics (Havana, Cuba)
Quantum Coherent Dissipation – a glimpse of the 'cat'

3/04 Seventh Symposium on Molecular Reaction Dynamics in Condensed Matter (Laguna Beach, CA)
Quantum Coherent Dissipation – a glimpse of the 'cat'

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